**Supporting Information**

**Interstrand side chain – side chain interactions in a double helical foldamer**

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**Mass spectrometry analysis of the single helix – double helix equilibrium.**

Maldi-TOF MS analysis of oligomers 1 and 2 only reveal a peak corresponding to the singly positively charged species of the monomeric helix. Presumably, the acidic matrix used for laser ionization disrupts the double helical complexes. On the other hand, Electrospray-MS analysis of dilute solutions support the idea of a monomer – dimmer equilibrium. As illustrated in Figure S1 for oligomer 2, the proportions between the two species is shifted in favor of the single helix upon increasing the proportion of MeOH in CH$_2$Cl$_2$ solutions at constant total concentration. This can conveniently be observed using a single peak where the isotopic distribution of the singly charged monomeric helix and doubly charged dimeric helix partly overlap.
Figure S1. Theoretical isotopic distribution of $[(2\text{-H})_2]^{2+}$ and $(2\text{-H})^+$, and experimental ESI-MS spectra showing the overlap of these distributions for 2 in solution in two different solvents. The double helix is the major species in pure CH$_2$Cl$_2$ while the single helix only slightly prevails in the presence of 90% MeOH.
DOSY NMR experiments showing the single helix – double helix equilibrium

Figure SI2. Calibration of DOSY experiments using an oligomer (structure shown on top) described in reference 10c for which the proportions between single helix and double helix are even in a 5 mM solution at 25°C in CDCl₃, and not broadened by exchange phenomena. The upper spectrum is a 1D 400 MHz ¹H NMR spectrum and the bottom is a 2D DOSY spectrum showing the clear distinction of the amide resonances of the single helices (at lower field) and of the double helices (at higher field).
Figure S13. NMR spectra of 2 at 25°C in DMSO-d$_6$. The upper spectrum is a 1D 400 MHz $^1$H NMR spectrum and the bottom is a 2D DOSY spectrum showing the distinction of the amide resonances of the single helices (at lower field) and of the double helices (at higher field). The resolution is not as good as in Figure S2 because of the broader lines of compound 2 in DMSO.